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METASTABLE STATES IN MEDIUM - AND HEAVY - WEIGHT NUCLEI

BY AUGUST ALEXANDER EBEL U. S. Naval Postgraduate School Annapolis, Md.









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August Alexander Whel B.A. Iowa State Toschers College (1962)

Submitted in Partial Fulfillment of the Requirements for the Degree of

DOCTOR OF PHILOSOPHY

at the

MASSACHUSETTO INSTITUTE OF TUCHROLOGY

(1950)



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ABSTRAGT

This thesis describes an experimental investigation of the emoitation of the metastable states in In²¹⁵ and Au²⁹⁷ by inclastic scattering of neutrons. The results are compared with those for quantum excitation of these states.

The neutron excitation curve for gold shows two pronounced discontinuities in slope at 1.1h ± 0.03 Nev and 1.hh ± 0.03 Nev, corresponding to energy levels that decay to the metastable state. The threshold for production of the isomer with neutrons is found to coincide with the energy of the metastable level.

It is shown that the spin of the notastable state must be at least 11/2. A comparison of the cross section for the direct production of this state with the cross section for formation of the compound mecleus indicates that the state is excited by an $\ell=3$ neutron. This fixes the spin at 11/2 with parity opposite to that of the ground state.

An analysis of quantum-excitation data shows the 1.14-lev level to have a spin of 5/2 or 7/2 also with parity opposite that of the ground state. From the properties of the isomeric transition, the spin of the known level between the metastable level and ground is shown to have a spin of 5/2 with parity opposite that of the ground state.

The curve obtained by neutron excitation of indium reveals a threshold at $600 \pm h0$ kev and other excited levels at $960 \pm h0$ kev and $1.37 \pm 0.0h$ Mev. The last two have been reported from quantum excitation at 1.0h and 1.42 MeV.

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Spin assignments of 1/2 for the netastable state and 3/2 or 5/2 for the 600-by level have been made. The parity of both levels is exposite to that of the ground state.

Confirmation is given to the existence of a beta-locay of the metastable state of In 115.

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I. INTRODUCTION

ORIGIN OF THE PROBLEM

Of special interest in nuclear physics is the determination of the energy, spin, and parity of excited levels in nuclei. With only a few exceptions, the lifetimes of these levels are too short to permit a direct measurement. Instead, their characteristics must be inferred from transitions to and from the ground state. Such studies involve nuclear reactions which leave the nucleus excited. The inclastic scattering of neutrons is one such reaction. In addition to its value in the theory of nuclear structure, inclastic scattering is of considerable practical importance in the problem of nuclear shielding.

cause of the small cross section and the inherently large background. In a few cases, however, the lifetime of the excited state is long enough to permit its measurement to be made after the beam of incident neutrons has been removed. Thus, neutron excitation of metastable states affords a very convenient method of investigating inelastic scattering in certain nuclei.

The purpose of this thesis is to investigate the inelastic scattering levels in Au¹⁹⁷ and In¹¹⁵ which decay to the metastable states of these isotopes. From an analysis of the neutron excitation curves, the spin and relative parity of many of these levels will be determined.

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HISTORY OF ISOMERISM

An isomeric state is generally defined to be an excited state of a nucleus which exists for a measurable length of time. It differs from other excited states only in the probability of decay to a lower state. With the recent advances in techniques of measurement, many states have been reclassified isomeric, and many more are being found.

The first case of isomerism was the discovery by Mahn¹ in 1921 that Pa^{23h} had two half-lives, one of 1.15 minutes and the other of 6.7 hours, associated with two beta-decay energies. He was able to show that both states grow out of the beta-decay of Th^{23h}. In spite of the search for other examples, it was not until the year after the discovery of artificial radioactivity in 193h that the next isomeric pair was found. Neutron, especially slow neutron, bombardment of bromine yielded three different radioactive decay constants, although bromine was known to have only two isotopes, Br⁷⁹ and Br⁸¹. It was shown that these three periods were due to capture of neutrons giving Br⁸⁰, Br⁸², and an isomer of the former.

Table I lists the lifetime and excitation energy of 23 known isomers of stable nuclei, together with the relative isotopic abundance of the stable state. Also included are the properties of the radiation arising from neutron capture which might interfere seriously with measurements of the neutron activation of the isomeric state.

With the existence of isomerism well established, the next step was a theoretical explanation by Weizsacker . He established that the assumption of an excited level of a few hundred key above the ground

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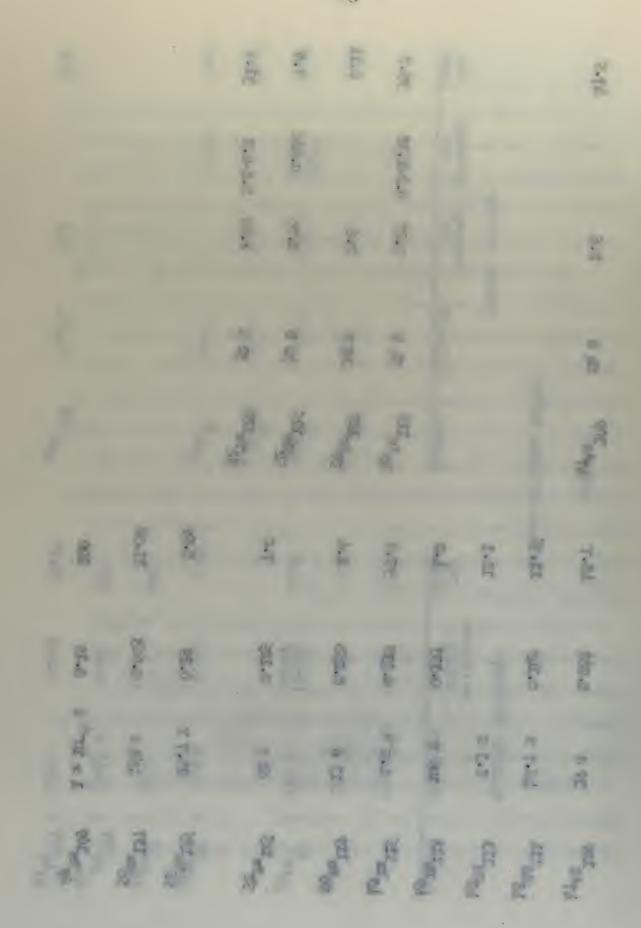
TABLE I

Isomers of Stable Melei

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capture activity, which saight confuse measurements on the isomer, is listed according to the stable inclose The data in this table are taken from tabulations by Segre and Selects and by Seeborg and Serland. The which captures the neutron.

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state with a spin difference several units was sufficient to predict such lifetimes.

It had long been suspected that an isomer is merely a metastable level, but the influence of atomic spectra in which large spin changes do not appear was carried over into nuclear considerations. Flugge showed this difference very clearly when he compared the atomic case having a Coulomb potential with a nuclear case assuming a potential well in a Fermi gas model. According to the build-up principle for electrons, the principal quantum numbers, n, and the azimuthal quantum numbers, n, are such that, although n gets very large, nowhere does n exceed three. In Flugge's hypothetical nuclear model, on the other hand, it is found that n may be four or five for protons and as high as six or seven for neutrons. This scheme also shows that isomerism is unlikely in light nuclei.

More recently layer has proposed a model based on strong spinorbit coupling which predicts the existence of large spin differences in low-lying levels. This explains in a rough way the "islands" of isomerism that have been observed in tabulations of isotopes. In many cases, however, the predicted spins and parity of the excited state do not agree with the experimentally determined values.

EXCITATION OF METASTABLE STATES

The excitation of metastable states may be accomplished in general by two methods:

1. Electromagnetic excitations by high-energy x-rays or the electric field of charged particles; and,

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old for neutron excitation, all determinations of the levels decaying to the metastable state have been made by the first method.

Excitation curves giving the energies of levels decaying to the isomer have been determined by x-rays for In¹¹⁵, Au¹⁹⁷, Rh¹⁰³, Cd¹¹¹, and Ag. Indium is described in Chapter V and gold in Chapter IV.

The other three isomers have been studied by Wiedenbeck^{8,9}.

Unfortunately, he found it necessary to use a thick target to produce x-ruys, so that the decision on the existence and location of the levels is subject to large error. Figure 1 shows the level diagrams that have been reported for these nuclei.

The two stable isotopes of silver, Ag¹⁰⁷ and Ag¹⁰⁹, have metastable states with very similar properties, the first having a half-life of h₀ seconds and an energy of 93 keV compared to 39 seconds and 63 keV for the second. It was therefore impossible to separate the levels belonging to one isotope from the other.

Neutron excitation by the (n,n) reaction to obtain an excitation curve has been reported for In¹¹⁵ by Cohen¹⁰ in the region above 2 Nev and at four points between 600 and 1500 kev by Taschek¹¹. Neither of these investigations shows the presence of levels other than the threshold level. Diedenbeck¹² has studied Au¹⁹⁷ in the vicinity of the photon threshold. However, no careful examination has been made of the neutron excitation function of any metastable state in the region from its threshold to 2 New with an attempt to locate the various different levels which decay to such a state.

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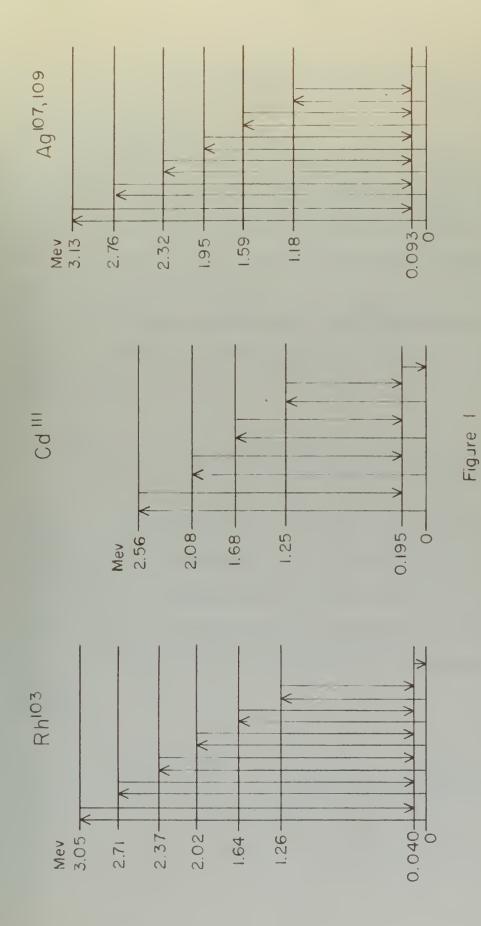
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ENERGY LEVEL DIAGRAMS for Rh¹⁰³, Cd ¹¹¹ and Ag^{107, 109} PROPOSED by (From Phys. Rev. 67, 267 (1945) and Phys. Rev. 67, 92 (1945)). WIEDENBECK from QUANTUM EXCITATION CURVES.



II. TWOR TICAL CO I ATIOMS

WINGSACHUR HYPOTHUSIS

A quantitative explanation of the lifetime of metastable states is complicated by the lack of detailed knowledge of the nucleus. As a result, there are many expressions for the calculation of the half-life of an isomeric state, depending on the assumption of the motion of charges in the nucleus.

From classical electrodynamics, considering an oscillating system of charges, the electric field at a point R, Ω may be expressed as 13

$$\underline{\mathbf{E}} = -\frac{1\omega}{c^2} \frac{e^{1\omega(t-R/c)}}{R} \int \underline{\mathbf{j}}_{\perp} e^{1\kappa \underline{\mathbf{n}} \cdot \underline{\mathbf{r}}} d\tau$$

where j_{\perp} is the projection of the current vector on the plane perpendicular to the unit vector, \underline{n} , in the direction of the point R, Ω , and r, θ are the coordinates of the charge producing the current. The integration is to be carried over the distribution of charges.

According to a well-known expansion, the exponential may be expressed as

$$e^{i k \, \underline{n} \cdot \underline{r}} = \sum_{\ell=0}^{\infty} \frac{i^{\ell} (k \, r)^{\ell}}{1.3.5...(2 \, \ell-1)} \, P_{\ell} \, (\cos \theta)$$

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in the case in which the wave length of the ridiation is long compared with the size of the charge distribution.

This leads to an expression for E as the sum of integrals of various powers of kr = r/k corresponding to the electric field of different 2 multipole orders. The first few terms are given below:

$$\underline{E} = -\frac{1\omega}{e^2R} e^{1\omega(t-R/c)} \left\{ \mathcal{L} \int_{\frac{1}{2}} P_0(\theta) d\tau \right\}$$

Consideration of the magnetic field laids to a similar result.

The transition probability may be computed by forming the Poynting vector and dividing by the energy of the radiation. Collecting the terms of like powers of kr gives an expansion for W., of the form:

$$W_{\gamma} - W_{E_1} + (W_{\gamma_1} + W_{E_2}) + (W_{W_2} + W_{E_3}) + \cdots$$

in which

$$W_{\ell-1} = W_{\ell} = (1/K)^{2\ell+1} = \frac{2}{5} n^2 \frac{(r/K)^{2\ell}}{\sqrt{1.3.5...(2\ell-1)J^2}}$$

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The approximation to the probability for gamma-decay of the isomeric state given above was based on Bethe's list assumption of a uniform-charge distribution throughout the nucleus. Various other models have been assumed for this derivation. Nebb and thlenbeck have assumed the radiation to be due to the electromagnetic field of a single alpha-particle moving in a potential well formed by the nucleus. Their result shows approximately the same dependence on the multipole order but includes a dependence on the charge of the nucleus. They give for the transition probability of the 2 lectric or 2 left agnetic radiation:

$$\lambda = 8(\frac{1}{2})(\frac{l+1}{l}) \frac{e^2}{2} (\frac{(r/k)^2 l}{2(1.3.5...(2l+1)^2)})$$

Flugge 16 does not consider the charge distribution performing a linear oscillation, but instead he pictures the radiation arising from a rotational excitation of a charged drop. This leads to another expression similar to others but differing in the constants arising from the nuclear properties and is of the form:

$$\lambda = z^2 \frac{e^2}{\pi c} \frac{\pi}{\theta} \frac{E}{Mc^2} (r/\pi)^2 \ell$$

where 0 is the moment of inertia and 1 the rest wass of the nucleus.

The accuracy of any of these formulae is sufficient to determine the ℓ value of the transition in most cases because of the vast dif-

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ference in lifetimes for different multipole-order transitions. However, an error of as much as 10² to 10² may be expected due to the inaccuracy of the particular model. None of these models appear at present to have much advantage over the others.

ANGULAR POMENTUM SELECTION RULE

By a consideration of the fields of the region in which they do not decay as R^{-1} close to the source, Heitler¹⁷ has shown that quantization of these fields results in the radiation of a 2^{ℓ} electric or magnetic pole having an angular momentum of ℓ in with respect to the radiation origin.

Since vector angular momentum of the nucleus must be conserved, the emission of radiation changes the angular momentum in accordance with the multipole order of the radiation. If the initial and final states of the nucleus differ by more than one unit of momentum in absolute magnitude, this places a lower limit on the order of the multipole. Conversely, if the lifetime of the excited state indicates that the lowest multipole order present is 2^{ℓ} , it can be stated that $|I| = |I| = \ell$.

Although 2 may be the lowest-order multipole radiation possible, higher-order radiation is of course also possible up to the limit determined by |I| + |I'|. However, barring a coincidence of charge distribution that reduces the strength of the 2 pole far below that which would be predicted from the size of the nucleus, only the lowest-order radiation would be observed because of its much greater probability.

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PARITY SELECTION RULA

Another selection rule that must always be obeyed is the conservation of parity in a transition. It can be shown by a consideration of the symmetry properties of the eigenfunction of the nucleus that emission of a $2^{\ell-1}$ magnetic or 2^{ℓ} electric radiation always involves a parity change for the nucleus if ℓ is odd and never if ℓ is even. Thus, although these transitions involve the same probability in the rough derivation above, it is impossible for both to occur between the same two states.

INTERNAL CONVERSION

Any comparison of these results with theory involves a correction for the competing reaction, internal conversion. Taylor and Nott lê have shown that internal conversion, being a one-step process and not a photoelectric effect of the outgoing gamma-ray on the atomic electrons, is in fact a competing reaction reducing the lifetime of the excited nucleus. Therefore, it is correct to define the internal-conversion coefficient as the ratio of the conversion electrons to the number of gamma-ray quanta and not as the ratio to their sum.

The calculation of these coefficients involves an evaluation of the probability of transmitting to the atomic electrons the excitation energy of the nucleus by a perturbation of their wave functions, this perturbation being caused by a multipole in the nucleus of such strength that it radiates one quantum per second. Since the fields due to the higher multipole orders decrease much more rapidly with distance than do

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those of the lower orders, this factor provides another check on the order of an observed transition.

Such a calculation in the general case is so complicated that the formulae derived are very restricted in their application. Dancoff and Eprison 19 have obtained expressions for the case in which the emitted electrons can be treated nonrelativistically and in the relativistic case in which the binding energy of the electrons in the shell can be neglected. However, numerical integration of the wave functions by Rose 20 shows that, even in this limited region, the predicted values may be in error by a factor of three or more especially for the higher-order multipoles and magnetic conversion. There are several other approximate formulae available, but these are no better when checked with Rose's data. Therefore, at the present time, all calculations of internal conversion coefficients must involve numerical integration of the Schrodinger equation.

The accurate experimental determination of the internal conversion coefficient is difficult because of the uncertainty of the relative efficiencies of the detectors for electrons and gamm-rays. The ratio of the coefficients for the K and L shells is much more accurately determined and is a more sensitive function of the transition, particularly in separating electric and magnetic transitions of similar probabilities. For example, gold has a measured K to L ratio of 3.4. Assuming electric 2⁴ transition gives 0.51 for this factor, and a magnetic 2³ pole has 4.8. The transition is therefore considered to be magnetic.

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And and Pancoff²¹ have compared the lifetime of 55 known isomers with the theoretical lifetimes predicted by the Weizsacker hypothesis corrected for internal conversion and find that these isomers do in fact fall into groups characteristic of $\mathcal{L}=1$ and $\mathcal{L}=5$ spin change, with one case, that of Pb²⁰¹, corresponding to $\mathcal{L}=6$. This agreement indicates that the hypothesis is valid.

NEUTRON EXCITATION OF METASTABLE LEVELS

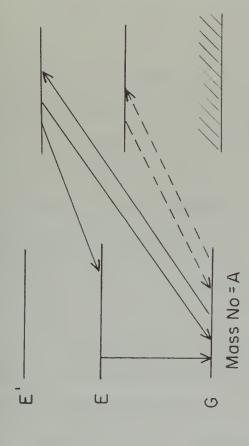
The neutron excitation of a metastable state is a special case of inelastic scattering of neutrons. The process is shown diagramatically in Figure 2. On the left is shown the bomburded nucleus with mass number A, and on the right is the compound nucleus with mass number A + 1 more tightly bound by the binding energy of the additional particle (on the order of 8 MeV). Mucleus A has a ground state 0 and a first excited level E.

Bosbardment by a neutron with energy less than the energy of level E is shown by the dotted line. In this case, the compound state can emit a neutron leaving the nucleus A only in the ground state. However, if the bosbarding neutron has a higher energy than does the excited level, the compound nucleus can decay to level E or to the ground state, as shown by the solid lines. The lowest neutron energy that excites level E is the energy of that level, provided the neutron is able to carry the difference in angular momentum between states 6 and E. If this is not the case, it may be necessary to excite a higher state E', whose angular momentum lies between that of E and G. Ievel E may then be excited by the emission of a gamma-ray by level E'.

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Region of overlapping energy levels and continuum.

G Mass No = A+I

SCHEMATIC DIAGRAM of NEUTRON SCATTERING with NEUTRON ENERGY LESS (broken lines) and GREATER (solid lines) THAN Figure 2 FIRST EXCITED LEVEL E.



The cross section for exciting level E is the product of the cross section of exciting the compound nucleus, o-c, and the probability, WE, of that nucleus decaying to level E.

Blatt and Teisskopf²² have derived an expression for the cross section for formation of the compound nucleus as a function of the energy and orbital momentum of the neutron with respect to the target nucleus. This is given to be:

where $\tilde{\tau}$ is the wave length of the incident neutron and T_{ℓ} is the transmission coefficient for the neutron through the "centrifugal" potential and also includes the reflection due to the energy discontinuity at the nuclear surface.

Assuming a square potential well for the nucleus, the transmission coefficient is shown to be:

for the case in which the wave number K of the neutron in the nucleus is large compared with the wave number k outside the nucleus. In this expression x = kR and X = kR, where R is the nuclear radius and v_{ℓ} is a factor determined by the wave function of the neutron beam at the surface of the nucleus.

The values of \mathbf{v}_ℓ may be calculated from a tabulation of Blatt and Weisskopf giving:

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$$v_0 = 1$$

$$v_1 = \frac{x^2}{(1 + x^2)}$$

$$v_2 = \frac{x^4}{(9 + 3x^2 + x^4)}$$

$$v_3 = \frac{x^6}{(225 + 1.5x^2 + (x^4 + x^6))}$$

$$v_i \rightarrow \frac{x^2}{(1.3.5...(2 - 1))^2}$$

The last expression gives the value of v_ℓ in the limit $x << \ell$.

It should be noted that the angular momentum a nautron can carry into the compound nucleus for a given energy is much greater than would be predicted from a simple quantization of the impact parameter. An 600-key neutron incident on Au^{197} can react only when it has orbital angular momentum $\ell=0$ or $\ell=1$, according to the naive approach. Actually, the cross section for $\ell=2$ is greater than for $\ell=0$ and over half that for $\ell=1$. $\ell=3$ is more than one-tenth and even $\ell=1$ is more than one-tenth and

The probability for decay to a given state from the compound mucleus follows from a consideration of the reciprocity theorem which states that

connects the cross section for a transition from state A to state B with the reverse transition. Reglecting all methods of decay of the compound nucleus other than neutron emission, the fraction of the compound nuclei which decays to state I with energy E₁ above the ground state in competition with a other states i of energy E₁ is given by:

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$$(z - z_1) \sigma_c(1)$$

$$(z - z_1) \sigma_c(1)$$

$$(z - z_1) \sigma_c(1)$$

where $\sigma_c^{(1)}$ is the cross section for the formation of the compound nucleus from level 1 and 2 is the energy available for the transition from the compound nucleus to the ground state of the product nucleus.

In a small energy interval just above the threshold for level "1" the sum in the denominator will change only slightly, so that in this interval the probability is proportional to $(E-E_1) \sigma_c^{(1)}$. As the energy of the compound nucleus is increased, the increase in the sum can no longer be neglected, and the probability will be less than predicted by the proportionality.

An inspection of the form of $\sigma_{\rm C}^{(\ell)}$ in the vicinity of the threshold shows that for the case $\ell=0$ the cross section $\sigma_{\rm C}^{(0)}$ decreases as $(E-E_{\rm th})^{-1/2}$; therefore, the probability of decay to that state increases as $(E-E_{\rm th})^{1/2}$. For $\ell=1$, for the neutron emitted in the decay, the probability increases as $(E-E_{\rm th})^{3/2}$ and in general as $(E-E_{\rm th})^{(\ell+1/2)}$. Thus, it is easily recognized if the decay to an excited state is accomplished by the emission of an $\ell=0$ neutron because the excitation curve will approach the threshold with a vertical tangent; whereas in cases of higher ℓ , the tangent will be horizontal.

This type of calculation applies directly to the case in which the compound nucleus decays to the metastable state itself. In many

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cases, however, this transition is so improbable because of spin changes involved that the compound nucleus decays to an excited state with an intermediate spin, and that excited state decays in turn to the metastable level. The cross section for production of the isomer must then be corrected considering the relative probability of decay from the excited state to the metastable state and the other available levels. For this, the calculations described earlier in this chapter according to the Teizsacker hypothesis may be used.

COMPARISON OF NEUTRON AND PHOTON EXCITATION

Neutron and photon excitation of levels above the ground state differ in two fundamental respects. Since a neutron is re-emitted in neutron excitation, it is not necessary that the neutron have exactly the energy of the level, whereas, in the case of photons, the excitation is a line absorption. Hence, for photons, it is necessary to use a broad spectrum energy source, and increasing the photon energy gives an isochrount of the source. This makes the shape of the excitation curve dependent upon the photon source and not upon the excitation process.

The second difference is found in consideration of the angular momentum that can be carried in to change the nuclear spin from that in the ground state to that in the excited state. A dipole radiation can contribute only one unit and a quadrupole only two units. Excitation by higher-order multipole radiation is so unlikely that it is not detectable in the ordinary investigations. The same rules that make the transition from an excited state to the ground level by high

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multipole radiation improbable hold for the case of exciting that state from the ground level by high-order multipole radiation. Therefore, it is impossible to excite a metastable state of moderate lifetime directly by photons.

However, neutrons may carry in several units of angular momentum depending upon their energy and the size of the target nucleus. In the case of Au¹⁹⁷, a 600-kev neutron can change the spin of the nucleus four units and is able to excite the metastable state directly. On the other hand, neutrons can also carry in small angular momentum changes with a high degree of probability, so that any state which can be excited by photons can also be excited by neutrons; and, in addition, neutrons can excite many states which cannot be excited by photons.

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III. EXPERIMENTAL DEUTPMENT

MUURON SOURCE

The neutrons used for activation of the metastable state were obtained from the Ma⁷(p,n)Re⁷ reaction, using protons from the Malata Rockefeller generator. This reaction was chosen because it has a comparatively large cross section and provides nearly monoenergetic neutrons.

Since it was desired to study isomeric production near the threshold for this process, it was desirable that the reaction be endoargic so that neutrons with low energies could be obtained. With a Q-value of -1.63 MeV, the Id (p,n) reaction satisfies this condition.

The presence of a lower-energy neutron group with relative intensity of about 10 percent has recently been reported²³ at 30h key below the main group. Since the excitation curve for the metastable state rises quite rapidly, this lower-energy group causes no ambiguity in the results. It is of interest that the Au¹⁹⁷⁰ excitation curve (Figure 13) shows a slight rise within the statistical error at about 130 key above threshold; this may be due to this effect, but the results are not conclusive.

The target was prepared by evaporating metallic lithium (92.6 percent Li⁷) on a 10-mil tentalum backing. To prevent the proton beam from boiling the lithium off, the target was rotated with the beam striking approximately one inch off center. An even coating was obtained by rotating the target while the lithium was being deposited.

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Thus, the problem of installing the target without the lithium's being oxidized was eliminated.

The thickness of the targets used was of the order of 15 to 20 kev, as measured by the conventional method²¹ of taking the thickness to be the energy between the neutron threshold and the first geometrical peak in the yield curve at 0 degrees.

FOC LIFEILER GENERATOR

The proton accelerator was an electrostatic generator originally used to accelerate electrons. During the past two years, this generator has been converted into a positive-ion accelerator by the Muclear Shielding Group. Unfortunately, the beam emerged in a room located under ground so that the scattered neutron flux is quite high. The target was located 35 inches from the analyzing magnet and only 39 inches from the concrete floor. In other directions, the situation was considerably more favorable. For the measurements being reported, however, scattered neutrons offered no problem because the exposures were unde in the region of high direct flux close to the target, and the scattered flux, being of lower energy, had a smaller cross section for interaction.

The beam was defined by crossed slits above the analyzing magnet and by horizontal slits beyond. Openings of 1 millimeter in the exit slits corresponded to an energy resolution of 0.1 percent in the proton beam. However, for the measurements on gold, it was necessary to open the slits wider to obtain sufficient neutron flux.

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The proton energies were determined by a generating voltaster indicating on a 6 inch fan-type microsumeter. This meter could be read to 0.1 of a division (about 5 kev in neutron energy), and measurements on the $\text{Li}^{7}(p,n)$ threshold indicated that it was reproducible within this amount over the period of a run. Over a period of several weeks, however, the calibration on the generating voltmeter changed by as much as several percent; therefore, it was always checked against the $\text{Li}^{7}(p,n)$ threshold (1.882 MeV) at the beginning and the end of each run.

In addition to this calibration point, the circuit has been checked for linearity using $\mathbb{H}^7(p_2,n)$, in which the proton is accelerated in a singly charged \mathbb{H}_2 molecule, giving a threshold at 3.7th Nev, twice the $\mathbb{H}^7(p,n)$ threshold, the $\mathbb{H}^3(p,n)$ with a threshold at 1.019 Nev, and the $\mathbb{G}^{13}(p,n)$ with a threshold at 3.236 Nev. It was found that the calibration of the meter against proton energy was linear in the region of interest, although the linear portion did not extrapolate to zero.

From the proton energy, the neutron energy can be calculated by application of conservation of momentum and energy. A tabulation of this calculation prepared by Willard²⁵ was used in this investigation.

LONG COUNTER

The neutron flux was measured, using a long counter of design similar to that of Hanson and McKibben²⁶. This counter, shown with dimensions in Figure 3, consists of a cylinder of paraffin shielded by cadmium surrounding a BF3 counter. The BF3 counter was 1 inch in diameter with an active volume 12 inches long. The filling was of 96 percent enriched BF3 at 50 cm. of mercury pressure.

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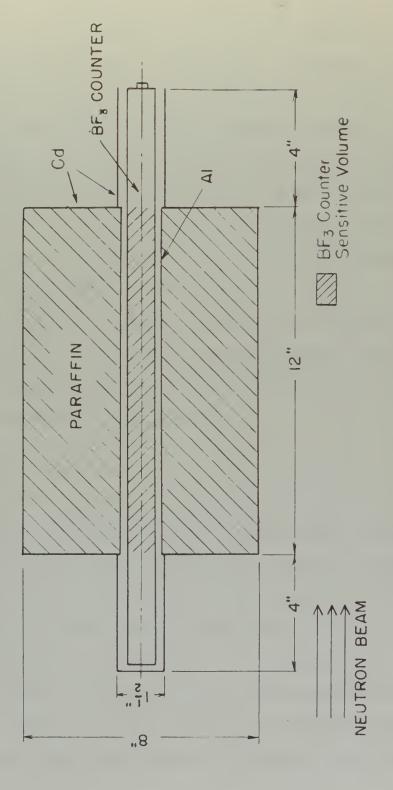


Figure 3. CROSS SECTION of LONG COUNTER



distincted by this lone counter with the curve published for a 10-by target by Enson, Taschek, and Tilliams²⁷. It assuming their yield curve to be correct, fillerd was able to determine a calibration for the long counter. Figure 1 show the relative response as a function of the energy of the incident neutrons. In addition, he found that, when located one mater from the target at 0 degrees, the counter gave 60 counts per microcoulomb of protons or key target thickness in the flat portion of the Li⁷(p,n) yield curve. The target thickness was measured by the initial rise in the yield curve. This gives an absolute calibration of the counter when command to Tanson's curve.

The associated electronics were of conventional design, consisting of a high-voltage supply, a model 100 linear amplifier, and two scale-of-6h scalars operated in series.

SCIPTILIATION COUNTRY

Decause it was hoped to take advantage of the difference in energy between the background and the radiation under investigation, a scintillation spectrometer was constructed. Although this discrimination did not prove feasible, the scintillation counter was used for the measurement of all induced isomeric transitions.

Figure 5 shows a block diagram of the counter setup.

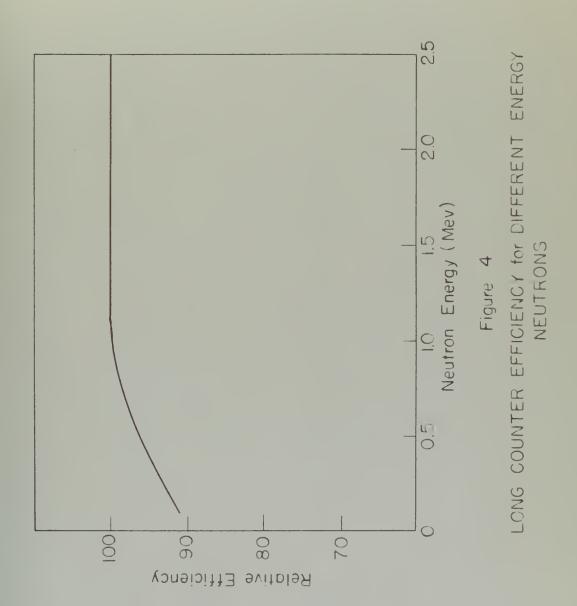
The counter itself was an RCA type 5819 phototube with an optically clear anthracene crystal 0.5 by 2 by 1; centimeters cemented to the tube window with Canada balsam. For increased light reflection, The first is the property of the second term of the property o

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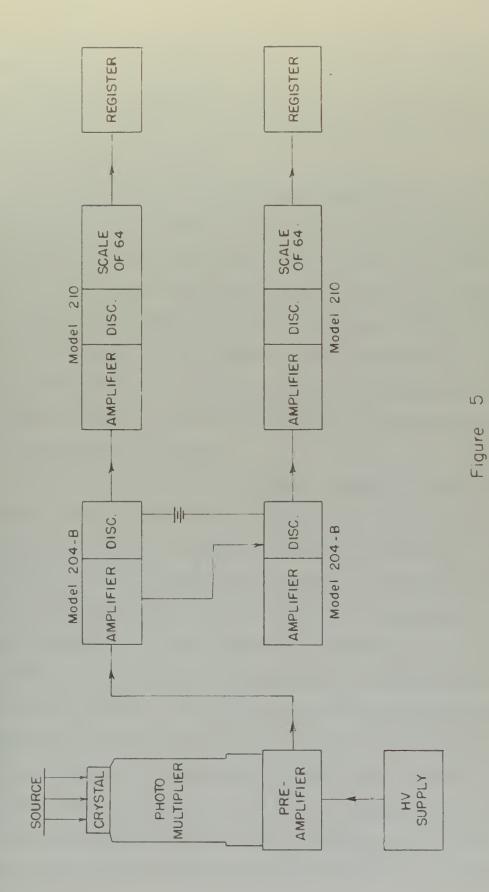
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SCINTILLATION BETA SPECTROMETER



the crystal was covered with thin aluminum foil (lair cm. thick). The tube and crystal were surround d on the side by a 0.010 inch mm metal whield for reducing stray magnetic fields and by 3 inches of lead to reduce the radiation background.

To permit rapid changing of someles, a brass plate containing a slide was fitted on the crystal end of the lead shield. The sample, in the form of a 3-cm. diameter foil, could be placed in the slide and moved to a point opposite the crystal about 3 millimeters from it.

Because of the critical dependence of phototube pulse-height output on the dyanode voltages, it is important that the high voltage be lept very constant. The high-voltage supply in the Atomic Instruments Nodel 105 Decade Scalar was used, and the voltage was monitored with an electrostatic voltagear.

The phototube signal was fed through a one-tube cathode follower preamplifier into an atomic Instrument Model 20hB linear amplifier.

This amplifier contains an integral pulse-height discriminator. Since the discriminator bias control furnished with the amplifier did not permit reproducible settings, it was replaced by a fifteen-turn Beckman heliopot. To obtain a differential discriminator, the integral discriminator from another 20hB amplifier was fed in parallel with the first discriminator. The voltage for the bias of the second discriminator was taken from the first with a mercury cell in series to provide the window. This system made the window error equal to the variation in the voltage of the series battery instead of the tracking error in two potentiometers. The two discriminators were fed into separate scalars, and the differential count obtained by subtraction.

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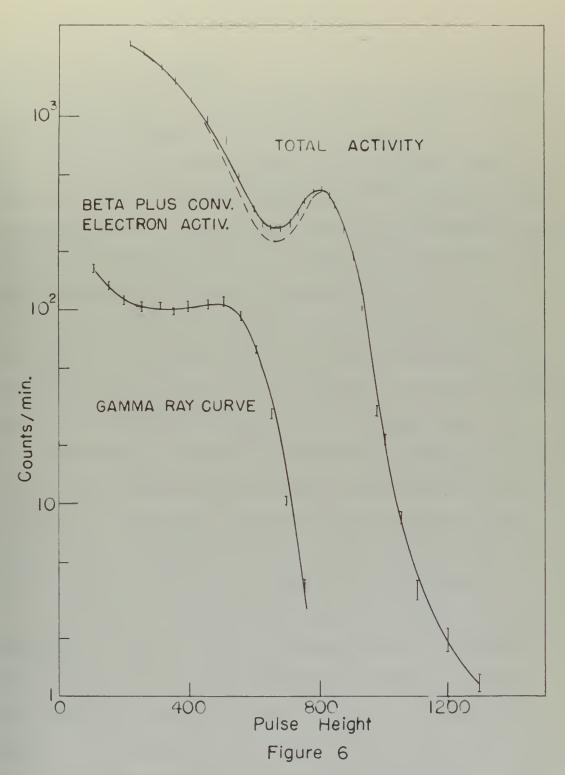
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Figure 6 shows the pulse-height distribution obtained from a Cs¹³⁷ source. The cesium activity contains a strong 30-kev conversion electron line which provided an excellent energy calibration point for the counter.

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Cs¹³⁷ DIFFERENTIAL PULSE HEIGHT DISTRIBUTION SHOWING 630 KeV CONV. ELECTRON LINE USED for ENERGY CALIBRATION.



IV. I.V TIGHT F COLD

HI TORY OF Au 197# EXCITATION

In an article surveying the field in 1941, Mattauch²³ observed that no cases of isomerism had been found for nuclei with a spin 1/2 < I < 9/2. This observation became known as "Mattauch's second rule." The first exception to be found was Au¹⁹⁷ with a spin in the ground state of 3/2. Later results indicate that the metastable state does not decay directly to the ground state, so that technically even this activity does not violate this empirical "rule."

isdenbeck²⁹ in 1915 reported a 7-second metastable state in gold excited by bombarding a gold cathode counter with x-rays. By using a thick gold target and aluminum absorbers, he was able to determine that the approximate energy of this level was 250 kev.

Subsequently, Wiedenbeck³⁰ investigated the quantum excitation curve for gold using the x-rays produced by monoenergetic electrons incident on a thick target. His results showed a series of levels starting with a threshold at 1.22 Nov and spaced about 0.1 Nev apart. The curve obtained and the levels assigned are shown in Figure 7.

In addition, he reported having excited this level by means of fast neutrons. Using a source of continuous x-r ys, he obtained a broad distribution of neutron energies from the $Be^9(\gamma,n)$ reaction. The experimental arrangement is shown in Figure 8. In the region of maximum neutron energy from 1.22 to 1.3 MeV, the counting rate in his gold counter was considerably above background and increased linearly with energy in much the same way as with x-rays. He concluded that the

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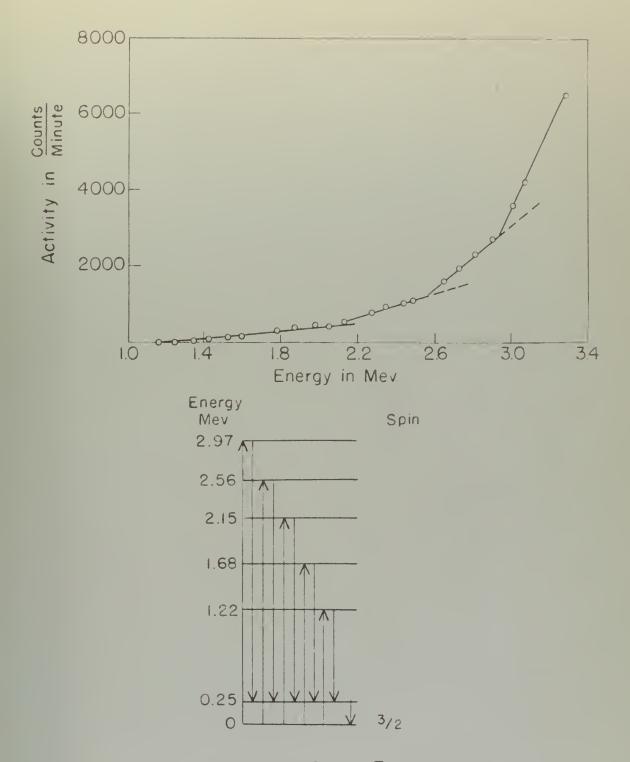


Figure 7
QUANTUM EXCITATION CURVE for Au^{197*} and LEVELS
OBTAINED by WIEDENBECK. (From Phys. Rev. <u>68</u>, I (1945)



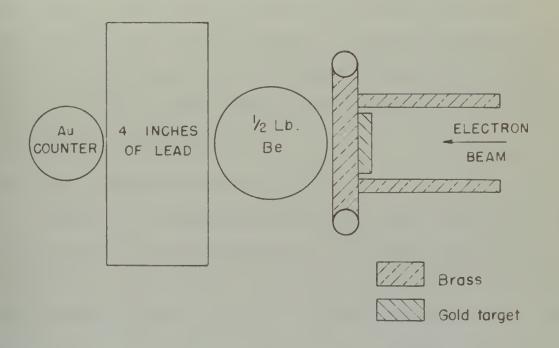


Figure 8

ARRANGEMENT USED by WIEDENBECK to IN-VESTIGATE NEUTRON EXCITATION of Au¹⁹⁷. (From Phys. Rev. <u>68</u>, I (1945)



threshold for neutron excitation was the same as for x-rys. His investigation, how ver, was limited to the energy int rval close to the photon threshold by a ximum neutron or ray available.

In the determination of the excitation function, using either quanta or particles having a continuous or ray distribution, the levels only appear as slight changes in the slope of a curve; hence, the nergy assignment and often the unitence of the level are uncertain and may even involve a subjective decision. If monoenerg the sources are used, the breaks in the curves are such sharper, and the results are considerably more certain.

DESCRIPTION OF THE THERRY HEY IS I Au 197

Because of the complexity of its excited states, the various levels in gold have been the subject of a great deal of nuclear investigation. On the other hand, gold has the great simplification of being monoisotopic with a mass of 197. Another advantage in the present investigation is the absence of any short-lived neutron capture reaction; the only one known leads to Au¹⁹⁸ with a 2.7-day half-life.

The levels in Au¹⁹⁷ fall into three apparently independent groups, with no evidence of transitions between any of the groups, as shown in Figure 9. The levels are shown in three groups to emphasize the improbability of transitions between the groups.

The first group consists of a level at 77 kev that is reached by K-capture from the lower 65-hour isomeric state of Hg¹⁹⁷. It has been determined that this 77-kev level does not have an appreciable lifetime.

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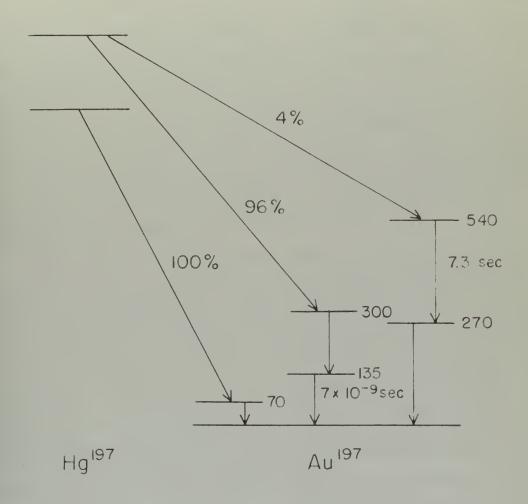


Figure 9

LEVELS in Au¹⁹⁷ as DETERMINED by the DECAY of Hg¹⁹⁷ (Energy in Kev)



The upper isomeric state in mercury with a half-life of 25 hours also decays by K-capture. In 96 percent of the cases, the decay is followed by the emission of a 165-kev game-ray to a 7 x 10⁻⁹ second isomeric level which in turn decays by the emission of a 135-kev game-ray^{32,33}. Assuming this transition to be electric quadrupole or magnetic dipole, the Weissecker hypothesis predicts a half-life for the game-transition of 6.5 x 10⁻⁹ seconds. This neglects the effect of internal conversion on the lifetime, but the calculations of Rose et al³⁶ indicate that the conversion coefficient will be less than unity and, hence, not sufficient to reduce the predicted half-life for a higher-order transition below a few microseconds.

The remaining I percent of the nuclei in the 25-hour mercury decays to a 7.3-second level which in turn decays by the emission of two internally converted games both of about 270 kev³⁵. Originally, the lower level was reported to be only of the order of 70 kev³⁶, and this value is found in much of the literature. This determination was made by means of an absorption of the conversion electrons in aluminum by Frauenfelder, Ruber et al. However, the reinvestigation by members of this group, using a magnetic spectrometer, revealed a coincidence rate in the K-conversion line for 270-kev games that was about twenty times the chance coincidence rate. The ratio of K- to L-conversion was also determined as 3.h.

Using the Weissacker calculation of the half-life with an energy of 270 kev, the following values are obtained for several different changes in medear angular momenta:

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TABLE II

Contractification	Parity Change	Half-Life (seconis)	Tlectric	la le pole
2	no	2.6 x 10 ⁻¹⁰	2-	21
3	yes	5.4 x 10-5	23	22
4	no	22.	Sft	23
5	705	1.6 x 107	25	Spt

The symbol \wedge is introduced to characterize an electric 2^{\wedge} or magnetic $2^{(\wedge-1)}$ transition, both having the same robability under the eigsacher hypothesis.

From this table, the transition from the 7-second state is evidently of the \wedge = h type and, hence, accompanied by no change in parity. However, it is not possible to determine from the half-life whether the transition is an electric 2^{h} note which involves a spin change of h or is a magnetic 2^{3} pole with a spin change of 3. The ambiguity is removed by the conversion coefficient data which show the transition to be of the magnetic type³⁷.

Frauenfelder 33 reports that the state to which this isomeric level decays has no appreciable lifetime, and Segre and Helmholtz 39 set this lifetime at less than 1 microsecond. Lat r in this section, it will be shown that this transition a most be of the $\triangle = 2$ type, because of parity consideration. In view of the crudeness of the theoretical calculations of lifetimes, the possibility cannot be ruled out that this is a $\triangle = 3$ transition with a predicted lifetime of 54 micro-

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seconds. The other possibility corresponds to an electric dipole transition which in general does not occur because of the smallness of the electric dipole moment.

Figure 10 shows the energy levels involving the 7-second motastable level, together with the parity and spin assignments that may be made from neutron excitation of the isomer.

Because the decay of the 7-second level directly to the ground state is not observed, although the energy is twice that for decay in cascade, this transition sust have a \(\text{value greater than } \). However, this level may be excited directly by neutrons, as is shown later in this chapter.

A comparison with theory of the exparimental cross section for the production of the isomeric state shows that the neutron cannot carry in more orbital angular momentum than l=3. The shape of the excitation curve shows that the neutron is emitted with l=0. To be added to this value of l=3 is the contribution of the neutron spin which may be -1, 0, or 1, depending upon the orientation of the spins of the incident and emitted neutrons relative to the orbital momentum. Thus, a l=3 neutron can excite a level with a spin differing from the ground state by 2, 3, or h units. This reaction gives a change in parity of the excited state due to the l value being odd for the incident neu row and even for the emitted neutron.

The spin changes of 2 or 3 between the ground and the excited state with a parity change both give a lifetime corresponding to \wedge =3 which is much too short. The only other possibility, the spin change h

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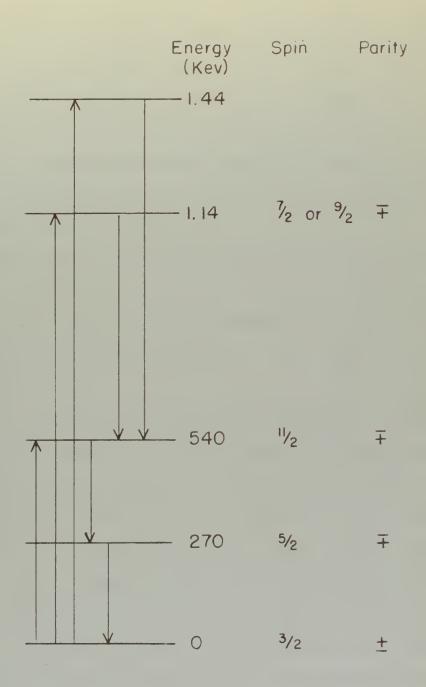


Figure IO

LEVELS EXCITED by NEUTRONS in Au¹⁹⁷ which

DECAY to the METASTABLE STATE at 540 KeV,



with a parity change, however, gives a magn tie 24-pole transition with a lifetime approximated by letting $\wedge = 5$. Thus, the spin and parity relationship between the ground state and the 7-second—tastable state are uniquely det rained, and it is shown that the neutron enters the Au¹⁹⁷ nucleus with orbital angular momentum of 3 and its spin parallel to the orbital axis, and it is emitted with angular momentum zero and its spin reversed.

Since the isomeric transition involves no parity change, the level to which it decays must also have a parity different from the ground state. This rules out the possibility of a transition from this state to the ground state having \wedge even, and in particular it excludes $\wedge = 2$.

Two other levels at 1.11 and 1.11 Nev are found that may be excited by fast neutrons of < 2-Nev energy. Newever, at these energies, the incident meutron may carry in even more angular momentum. At the same time, there is no lifetime requirement on the state, except that transitions to the metastable state are quite probable. Furthermore, the shape of the excitation curve for each level individually cannot be determined because of the uncertainty in the contribution of lower levels to the isomeric state. For these reasons, little can be deduced from the neutron excitation curve about these states other than their energy.

It is of interest to compare the energies determined by neutron excitation with those Tiedenbeck has reported for quantum excitation. In Chapter II it has been shown that any level excited by a quantum should be excited to an appreciable degree by neutrons. The level

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excited by 1.1h-lbv neutrons appear ntly corresponds to the threshold found for 1.22-Vev quanta. The agreement is considered satisfactory because of the difficulty of deducing this threshold accurately from the small slope between the quantum excitation curve and the axis. However, no evidence is found with neutrons of the level reported for quanta at 1.68 lbv.

The fact that the 1.11-Mev level can be excited by quanta enables a determination to be made of its spin relative to the ground state.

The spin change of h from the ground state to the metastable state with a parity change must be accomplished by the absorption of a quantum plus the emission of another.

There are two possibilities with high enough probabilities for the process to occur. The excitation is either by an $\ell=2$ yes (magnetic quadrupole) radiation followed by decay to the isomeric state by $\ell=2$ no (electric quadrupole) radiation or excitation by $\ell=3$ yes (electric octupole) radiation followed by $\ell=1$ no (magnetic dipole) radiation. It is impossible to separate these since they both have the same probability according to the reizsacker hypothesis. However, the decision as to whether a transition corresponds to the excitation of the higher level or the decay to the netastable level is unambiguous because the decay to the metastable level must be able to complete successfully with the decay to the ground state in spite of the higher energy involved in the latter.

Unfortunately, the Wiedenbeck date do not permit even an inference regarding the probability of exciting the neutron level at 1.bl; liv with quanta.

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INJURIETION OF EMPERIMENTAL SETUP

Measurement of the neutron excitation function of Au^{197*} was complicated by two factors:

- 1. The cross section for the excitation process is so small that it was necessary to sacrifice part of the energy resolution to obtain activities large command to the background; and,
- 2. The half-life of the isomeric state is only 7 seconds. Figure 11 shows the experimental setup used for the gold investigation.

The technique used involved irradiating the sample in the form of a foil with a known flux of monoenergetic neutrons for 15 seconds, cutting off the neutron beam, transferring the foil to a scintillation counter, and counting the activity for 17 seconds.

The gold foil was 3 cm. In diameter and 0.1 mm. thick with a mass of 1.120 grams. To permit rapid transfer of the foil from the irradiating position to the counting position, the foil was mounted in a depression at one end of a balalite slide 1/8 inch thick by 1-1/2 inches wide by 3 feet in length. This slide was carried in a slot in the shield of the scintillation counter, which has been described previously. When the slide was against the stop at one end, the foil was accurately positioned must to the crystal of the counter. Then it was against the stop at the other end, the slide was held by aluminum fingers in front of the been to eliminate errors due to the flexibility of the slide. This arrangement was necessary to keep the counter out of the region of high neutron flux which would lead to high background counting rates for

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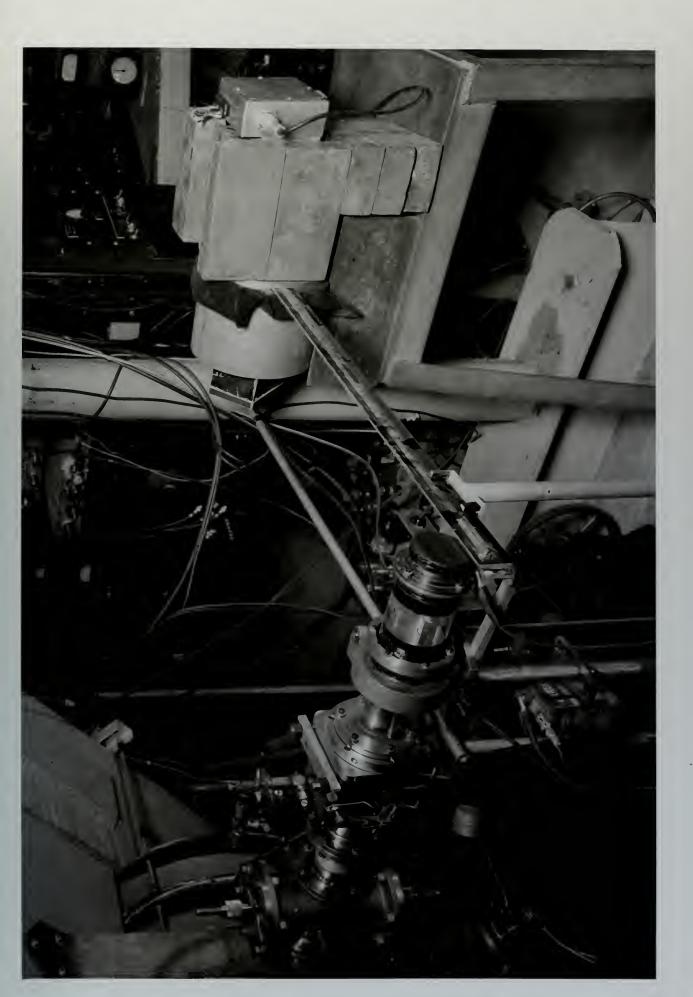
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Figure 11. Arrangement for irradiating the gold foils, showing the balkelite slide and the two counters with their lead shields.

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a short time after the beam was cut off. It also permitted shielding of the counter on all sides from the stray property background that was present even when the beam was not striking the target.

During the counting periods, the neutron beam was cut off by closing a tantalum-backed flap valve in the proton beam. This valve, unfortunately, was located near the target instead of at the entrance slit of the deflection chamber. For this reason, slight wanderings resulted in the beam striking the chamber walls which caused large fluctuations in the background during the counting period.

another scintillation count r with characteristics as nearly as possible like the counter for the gold was set up. This no occurrer was placed beside the other with the same amount of shielding but also was shielded from the gold activity. The scalars for both counters were arranged so that they were controlled by the same switch. Test activation runs were then made which were identical to the true runs, except for the omission of the gold from the slide. These runs showed the backgrounds of the two counters under those conditions to be the same within a statistical uncertainty of about 5 percent. Since the background over most of the curve was of the order of 10 percent or less of the induced activity, the matching was considered satisfactory.

The neutron flux for these runs was determined by the long counter previously described. The counter was set up one meter from the target at zero degrees. The number of counts during each irradiation was recorded and corrected for the effect of neutron energy on the counter efficiency.

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The exposure of the foil was initiated by the operator's pulling a cord with his left hand which opened the flap valve and allowed
the proton beam to strike the target. After I' second, the cord was
released and the flap was closed by a counterweight. At the same
time, the operator pulled a second cord with his right hand which moved
the slide into position, and with his left hand, he started the counter. This arrangement prevented the accidental starting of the counter
before the beam was off. With practice a rhythm was established that
permitted starting the counting priod one-half second after the end
of the irradiation period with a variation of less than one-fourth
of a second. This variation corresponds to a random error of less
than 3 percent.

TREATLESHI OF THE DATA

Each observation yielded on the order of 150 to 100 counts in the 15-second counting interval with a background of about 20 counts. At each energy, five or more identical runs were made and averaged to reduce the statistical and other random errors, such as variations in the length of time between the end of the irradiation and the beginning of the counting. An analysis of the individual readings taken at the same energy shows that they agree from the serves within the statistical error. The nonexistence of appreciable errors due to slow variations was shown by the reproducibility of the data over a period of several days.

Two tests were made to check that the 7-second isomeric state of gold was being counted. A decay curve of the radiation was taken by

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The selection determined decreased sold should stock only other press whench every bill section man are all the service speech in whethere probed new hilling moting the number of counts at the end of each 5 seconds. This method was subject to large statistical and human errors, but the results were consistent with a half-life between 5 and 10 seconds. Dince there are no other known activities which could be excited in gold with this half-life and since the activity was not found when the gold was removed from the slide, this was considered sufficient to identify the activity.

In addition, a pulse-height distribution curve as run to identify the energy. This curve also suffered from poor statistics but was consistent with the known charact ristics of the isomeric state.

In the determination of the excitation curve, the energy resolution was affected by three major factors, the fluctuations in the energy of the proton beam, the thickness of the lithium target, and the dependence of the neutron energy on the angle of emission. A fourth factor, scattered neutrons, may be neglected because of their low intensity relative to the direct beam in the region in which the foil was exposed.

To increase the beam current striking the target, it was necessary to operate with the beam defining slits opened to 5 millimeters. This leads to a resolution of approximately 15 keV in the proton beam and, hence, in the neutron beam. Since the beam striking the slits was large compared to the opening, all energies in this region are approximately equally represented.

The effect of using a target of finite thickness is to give also a square number-energy distribution for the neutrons similar to that

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caused by the slit opening. The effects are therefore additive and, according to measurements on the rise of the neutron yield curve at threshold, amount to a resolution of 30 keV in this investigation.

The third factor in the resolution, the angle at the target intercepted by the foil, must be considered because of the dependence of the neutron energy on angle. For these experiments in which the cross section for the reaction is quite small, it was found nace eary to sacrifice angular and, hence, energy resolution in order to place the foils close enough to get sufficient activity. Accordingly, the foils were exposed 5 centimeters from the target. They intercepted a half angle of 17 degrees for the pertion of the curve above 1.0 MeV. Below 1.4 MeV the exposure was made at h c ntimeters with a half angle of 21 degrees. This corresponds to an energy sweed of 25 keV at 600 keV in the forward direction and to 35 keV at 2.0 MeV.

Figure 12 shows the correction that must be applied to the maximum neutron energy to obtain the mean energy of neutrons incident on the foil. This curve was obtained by the following considerations.

For the purpose of calculating the mean energy incident on the foil, the beam may be considered uniform in intensity over the angle involved since the energies involved are well above the Li(p,n) threshold.

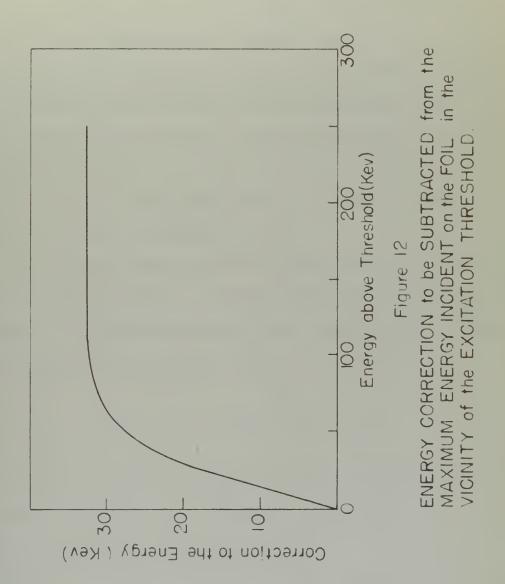
Let the energy spread due to the slit width plus the target thickness be represented by ΔE_p and that due to the finite solid angle intercepted be ΔE_q .

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The variation of the cross section for the production of the isomeric state may be considered constant over the energy served ΔE_0 except at threshold. Sense for the mount that the neutron bear is monoenergetic at 0 degrees; that is, that ΔE_0 is zero. Then, the activity due to each increment of energy is proportional to the area of the target subtended by the corresponding increment of angle at the source. This obviously increases linearly with energy up to the angle intercepted by the whole target feil. The mean energy in terms of the activity it produces is then calculated to be 0.707 of the total spread, ΔE_0 , below the 0 degrees energy.

This result may now be integrated over the spread of neutron energies present at O degrees. Again assuming the cross section to be constant over the energies involved, the correction to be subtracted from the maximum energy is found to be:

The calculation of course does not hold at threshold because of the assumptions on the effect of energy on cross section. Then the maximum energy is just equal to the turnshold for excitation, the correction must be zero because that is the only energy causing excitation.

Another point may be calculated by considering the case in which the lowest energy present is just the threshold energy. Again, with somewhat less justification, assume the cross section uniform over each Δ Eq. but this time assume that the cross section over Δ Eq. increases

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as $(E - E_{th})^{1/2}$. A calculation similar to that preceding gives a correction equal to:

This correction is somewhat too large due to the assumption on Δ but gives a reasonable amproximation of the true value.

The activity of the gold above background was divided by the neutron flux and is plotted in Figure 13 as a function of the mean energy of the neutron beam as corrected according to the preceding considerations. The upper portion of the curve talen with the foil 5 centimeters from the target was normalised to the activity expected at h centimeters to join with the lower portion of the curve. The two sections overlap in the energy interval from 1.0 to 1.4 MeV.

The neutron excitation curve shows the characteristic shape for times levels at 530 ± 20 kev; 1.1h ± 0.03 kev; and 1.1h ± 0.03 kev with a possible fourth level at about 1.77 kev. The errors assigned are estimates of the uncertainty in the determination of the energy of the neutrons and the location of the level on the curve.

The section of the curve in the vicinity of the 1.hl-May level was checked with higher resolution to be sure that the level was actually present. It was found that the break in the curve was readily reproducible.

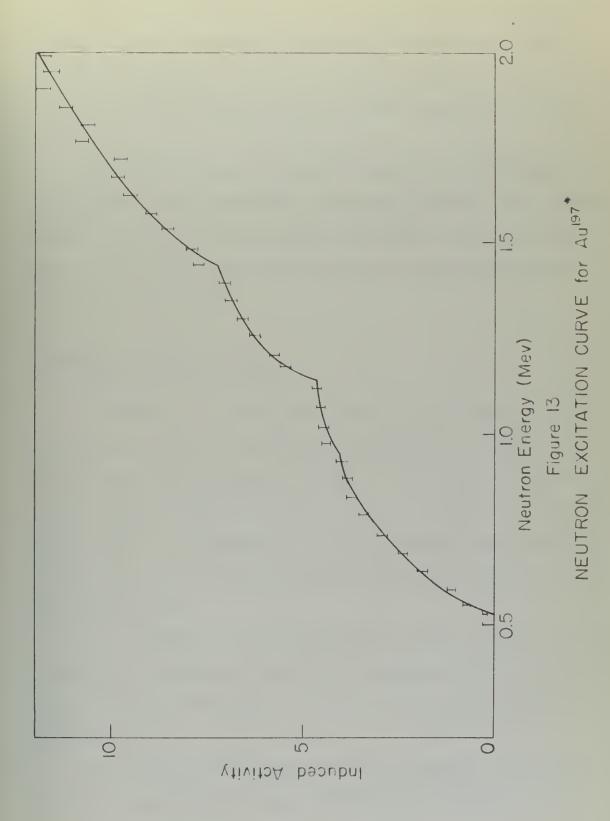
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The curve shows about a 10 present rise 130 kev above threshold. Although this may be explained by statistical uncertainty, it seems more probable that this is due to the second energy group reported to be 384 kev below the main group with a relative intensity of 10 percent.

The threshold value is of special interest because it corresponds quite closely with Ruber's measurement which shows the metastable level to be about 51:0 kev above the ground state. As a result, there appears to be little doubt that the neutrons are able to excite the metastable state directly.

COMPARISON OF THE THEORYTICAL AND EXPERIENTAL GROSS SOLICE

A knowledge of the cross section for the formation of the metastable state is of great importance in assigning an ℓ value to the incident neutron that excited that state. A comparison of this cross section with the $\sigma_c^{(\ell)}$ cross section for the formation of the compound nucleus Λu^{198} as a function of ℓ for the incoming neutron is sufficient in this case to place a maximum on the engular mountum the neutron can carry in.

The calculation of $\sigma_c^{(l)}$ following the without of Blatt and Weisskopf, as described in Chapter II, is carried out in the energy region of 600 to 1200 key for $0 \le l \le l_1$. The results are plotted in Figure 11.

Determination of the experimental cross section involves knowledge of the neutron flux on the gold foil and the efficiency of the

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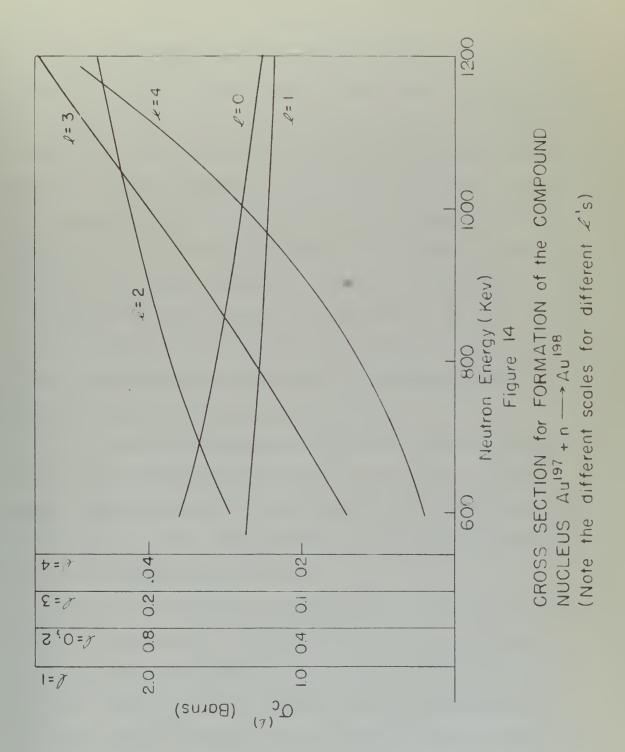
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counter for counting the excited state. The largest error in this cross section will result from the difficulty in evaluating the second factor.

In the data plotted on Figur 13, the ordinate is the number of counts of the Au^{197*} activity divided by the number of register counts from the long counter when operating through a scale of 102h scalar.

illard¹⁰ has determined that the flat part of the neutron yield curve this counter at 0 degrees and one ofter from the target gives 60 counts per microcoulomb of protons per key of target thickness, the target thickness being measured by the initial rise of the neutron yield curve with good proton energy resolution. From the data of lanson, Taschek, and illiams¹¹ on neutron yield, a ho-key target in this energy region gives h.35 x 10⁶ neutrons per microcoulomb per unit solid angle at 0 degrees. Therefore, each count on the long counter corresponds to

When exposed at 1: centimeters, the 3-centimeter foil intercepts a solid angle of 0.128 steradians. Hence, each count on the long counter indicates that the foil has been traversed by 232 neutrons; or, taking into account the scaling factor, each register count stands for 2.38 x 10⁵ neutrons through the foil.

The area of the foil is 7.07 square centimeters, and the mass is 1.120 grams corresponding to 1.31×10^{21} Au¹⁹⁷ nuclei. Therefore, an ordinate of unity on the plot in Figure 13 is equivalent to a cross section of 0.0069 barns if it is assumed that the scintillation counter counts every gold nucleus excited. This, then, is the minimal value

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that the cross section can have. On Figure 15, the minimal cross section for the production of the metastable state in the vicinity of the threshold is plotted as a function of energy, together with the cross section for the formation of the Au^{198} compound nucleus by an $\mathcal{L}=h$ neutron, $\sigma_{\mathrm{c}}^{(h)}$. It is seen that $\sigma_{\mathrm{c}}^{(h)}$ is even 1 so than this minimum over part of the region. This reaction can therefore be definitely ruled out as the method of empiting the 7-second state directly.

of course, the minimal value computed is too low by a factor equal to the reciprocal of the overall efficiency of the counter for counting the metastable state. Since the foil thickness is over 10 times the range of the conversion electrons, their contribution to the efficiency may be neglected, and the counting rate may be considered to come entirely from the gold x-rays produced on their emission and from the unconverted game-rays. Due to the resolution of the scalar following the counter, the decay of the nonisomeric 270-low state cannot be separated from the isomeric state.

Using the mass-absorption coefficient for the gold K-series x-rays of 3.40 cm²/gm, it can be calculated that in the one-dimensional case of a source 0.1 millimeters thick, the self-absorption is 0.73. The attenuation of this radiation in the crystal may be calculated to be 0.76, corresponding to an absorption of 0.24.

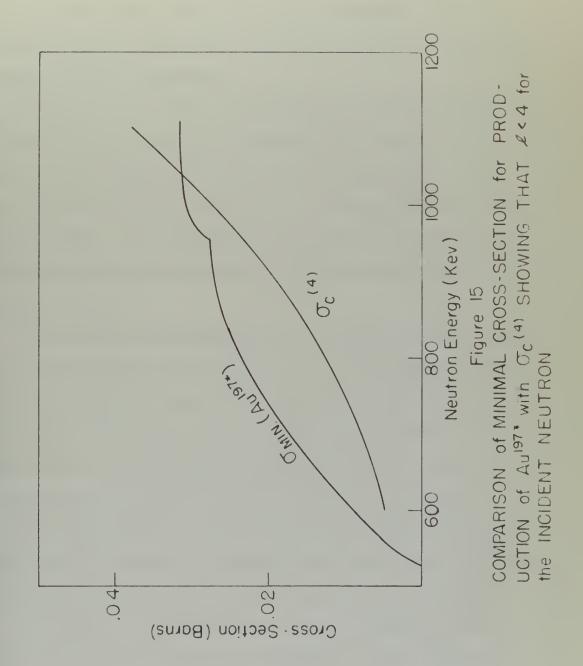
For the 270-kev unconverted radiation, the self-absorption is small and the crystal absorption 0.20. This gives an efficiency for counting the x-rays from this source of about 0.18 and for the game-rays about the same figure. So far, it has been neglected that only

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half of the radiation is directed toward the counter and that two decays take place for each metastable nucleus. However, these factors will careal each other to the extent of the uncertainty involved in this calculation. Softer x-rays, notably the I-series, have also been neglected in this approximation.

There is another factor to be considered, the afficiency of the photomultiplier for counting scintillations in the crystal. This has been checked by comparing the counting rate in this counter from a thin C^{lli} source with that obtained in a 50 percent geometry windowless gas-flow counter. It is found that this factor, which also corrects for the scintillation counter geometry being slightly less than 50 percent, is 0.90.

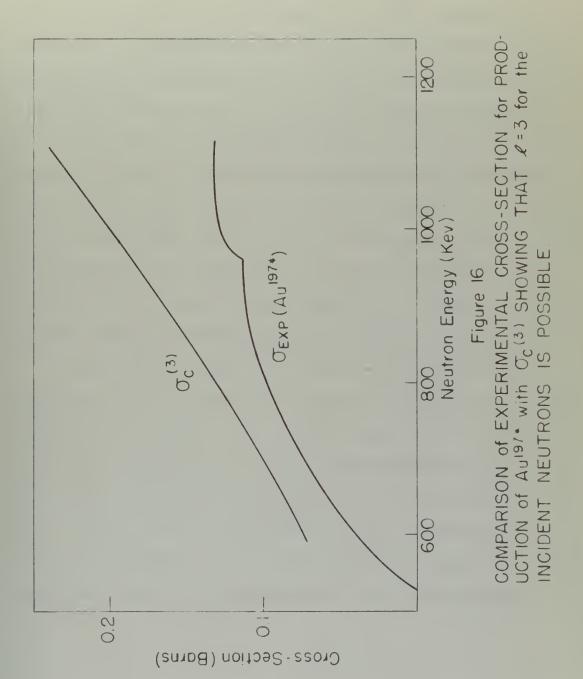
From all these considerations, it is estimated that the scintillation counter counts about 16 percent of the Au^{197*} nuclei decaying during the counting interval. The bias on the discriminator, however, was set so that 10 percent of the counts were not recorded, and the counting period of two half-lives was such that only 75 percent of the nuclei decayed during the period. Therefore, the correction to the cross section should be increased by a factor 1.45, giving a final value of 0.24. Because of the necessary crudeness of this calculation, it is considered that it may be in error by as much as 50 percent.

In Figure 16, the cross section for the formation of the metastable state in the vicinity of the thr shold is plotted with this factor applied to the minimal cross section. A plot of the cross section for the formation of the compound nucleus by an $\ell=3$ neutron is to the state of the contraction to state of the contract and this less decrease that of the state of the contract and the c

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also shown on the sam graph.

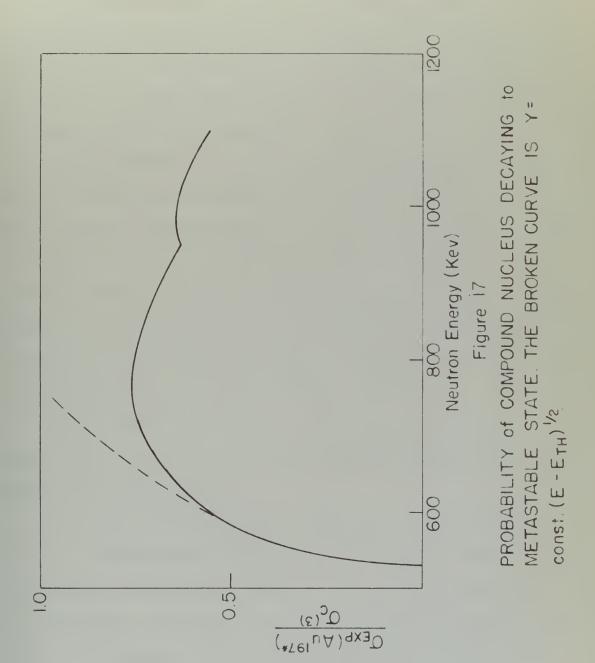
It is expected that the cross section for production of the isomeric state ould be smaller compared to $\sigma_{\rm c}^{(3)}$. Resever, the transition to the metastable state can compete very successfully with the transition to the ground state or other excited levels because it involves the emission of an $\ell=0$ neutron. Becay of the compound nucleus to the ground state requires that an $\ell=3$ neutron be emitted. Assuming that only the ground state and the metastable state compete, a calculation, such as described in Chapter II, reveals that 75 percent of the compound nuclei will go to the metastable level. Recalling that the experimental cross section may be in error by 50 percent in absolute magnitude, it is seen that the results are in agree ant with the assumption that the activation takes place with an $\ell=3$ neutron.

Figure 17 shows a plot of the neutron excitation curve from threshold to the first excited state above it, corrected for the cross section for the production of the compound nucleus. The resulting curve is the probability of emitting a neutron from the compound nucleus with L=0 and the spin opposite to that of the incident neutron. This curve has the typical shape for the emission curve of an L=0 neutron starting cut as an $(T-E_{\rm th})^{1/2}$ curve (also shown) and then falling below due to competition with decay to other levels.

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Figure 17 shows a play of the mentions empirical source from the extended to the first and obtained the common the common to the common for the common for





V. INVESTIGATION OF INDIUN

HISTORY OF In 115* WICHATION

The metastable state in In 115 has been studied by numerous investigators, not only to determine the properties of the state itself, but also as a convenient threshold detector. The relatively large cross section for production of the isomer and its convenient half-life make it ideal for many problems.

In 1939, Goldhaber, Hill, and Ssilard reported the excitation of In115* by inclustic scattering of neutrons from a radon-alphaberyllium source giving n utrons of a maximum energy of about 13.5 Mev. The results were checked by exciting the state with 2.5-1 or d-i neutrons. It was shown that the activity belonged to the mass 115 indium isotope and not to the mass 113 by producing Cd from Cd by an (n,2n) reaction. The Cd they decayed to In115*. On the other hand, Cd is stable and, hence, could not lead to In113*.

The metastable state has since been produced by Barnes and Aradine 13 with 6.7-1 ev protons and by Lark-Morovitz, Risser, and Smith 16-1 ev alpha-particles.

The first extensive excitation curve for this level was obtained by aldman and Miedenbeck . Using m-rays to produce the isomer, they reported levels at 1.12, 1.55, 2.13, and 2.64 Mev which decay to the notastable level. Subsequent masurements by Miller and aldman have confirmed the existence of the first two levels but corrected their energy to 1.04 and 1.12 Mev. The work by Maldman and Wiedenbeck re-

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quired the use of a thick x-ray target and therefore suffered from the same difficulty in interpretation as did diedenbeck's work on gold.

However, Miller and aldman found it possible to use a thin target that gave much more pronounced breaks in the excitation curve corr sponding to the levels. In addition, they found a weak activity (twice the background) with 857-kev x-rays which was considered to be possibly the excitation of an 873-kev level, reported by lawson and Cork 17.

A neutron excitation curve has been obtained by Cohantis using C¹³(d,n) and d-d neutrons from a 1-lev Cavendish generator. Because of the reaction that was necessary to produce the neutrons, the curve could not be carried to the throshold of the excitation but had to be stopped at about 2 Nev. Figure 10 shows the results that were obtained. It is to be noted that the presence of individual levels was not detected. This is probably because so many levels contribute to the excitation at the high energy involved that the breaks due to the individual levels could not be seen.

In connection with an investigation of threshold detectors, Taschek¹⁹ obtained four points on the neutron excitation curve in the energy region from 600 to 1500 kev. Of course, these data are too sketchy to show more than the general trend of the curve.

DESCRIPTION OF EMERGY LEVELS IN In 12

The energy of the isomeric state has been measured by Pell, Ketelle, and Cassidy using both a scintillation counter and a magnetic lens spectrometer. It is found that the F-conversion electrons have an energy of 312 keV, corresponding to an energy of 310 keV for the level.

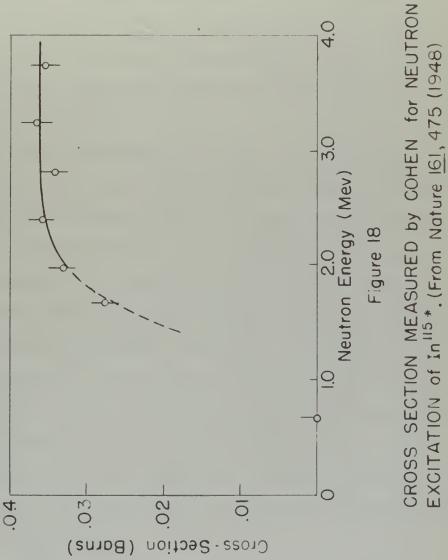
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The half-life of the state, according to the determination of lawson and Cork⁵¹, is h.5 hours.

Assuming the transition to be of the \wedge = k type, \wedge defined in Chapter IV, the Teizsacker hypothesis predicts a half-life of k seconds; while, if it is assumed to be \wedge = 5, the result is 800 hours. Although the agreement is not too good, it seems highly probable that the transition is of the latter type involving an electric 2^5 pole or a magnitude 2^k pole radiation.

The K- to L-conversion ratio in this case does not give a conclusive decision between these two possibilities. Amel and Danceff⁵² report a theoretical value of 6.8 for the magnetic case and 2.6 for the electric multipole transition. The experimental value given is 5.0. Thus, it would seem to favor choosing the egnetic transition.

Consideration of the spins involved, however, removes the ambiguity. The ground state has a spin of 9/2. The electric 2^5 pole transition requires a spin change of 5 between the ground and metastable states. If the possibility of a spin of 19/2 is ruled out because it is too large, there is no possibility for an $\mathcal{L}=5$ transition. On the other hand, assuming the metastable level to have a spin of 1/2 agrees with a magnetic 2^{14} pole transition having a spin change of 1 and a parity change.

Figure 19 shows the metastable levels, together with those found at 600, 960, and 1370 kev from the neutron excitation curve determined in this investigation. Also shown is the competitive decay of the metastable level in about 5 percent of the cases by emission of a

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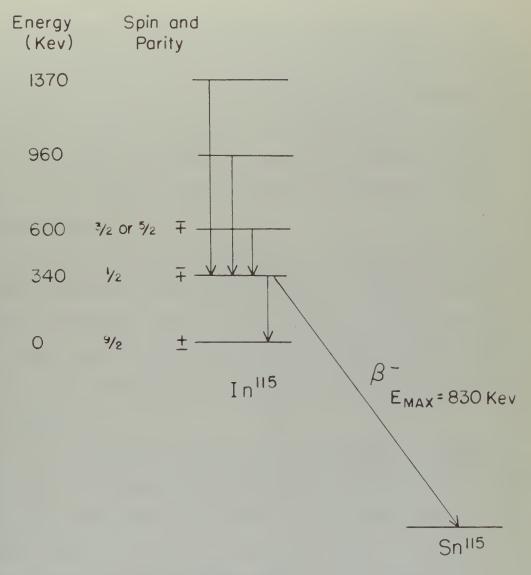


Figure 19

ENERGY LEVELS MEASURED in In 115 by NEUTRON EXCITATION of the METASTABLE LEVEL.



bota-particle having a maximum energy of 830 keV, as found by Bell, Ketelle, and Cassidy⁵³.

The existence of this beta-decay has been confirmed in connection with this study of indium. Figure 20 shows a pulse-height distribution obtained from an 8-mil indium foil bombarded with neutrons from the N. I. T. cyclotrom. The shape of the curve is distorted because the foil was thick compared with the range of the electrons, but the existence of the 312-kev conversion electron and 630-kev beta-decay is clearly shown.

It seems likely that the quantum-induced activity below 900 kev reported by Miller and aldman and by lawson and Cork was actually exciting of the 600-kev level. If this is assumed, it follows that the spin and parity of that level must be such that this process can take place often enough to be observed but that the process is somewhat forbidden.

A \(= 2 \) or lower transition (electric quadrupole or magnetic dipole) may be ruled out because this makes the transition to the ground state so much more probable than that to the metastable state that excitation would not be observed. On the other hand, \(\lambda = \) is a higher can be eliminated for the transition from the ground state to the 600-level because such a transition is too improbable for quantum excitation.

The remaining possibility $\Lambda = 3$ involves a parity change and a spin change of 3 for electric 2^3 pole or of 2 for magnetic 2^2 pole

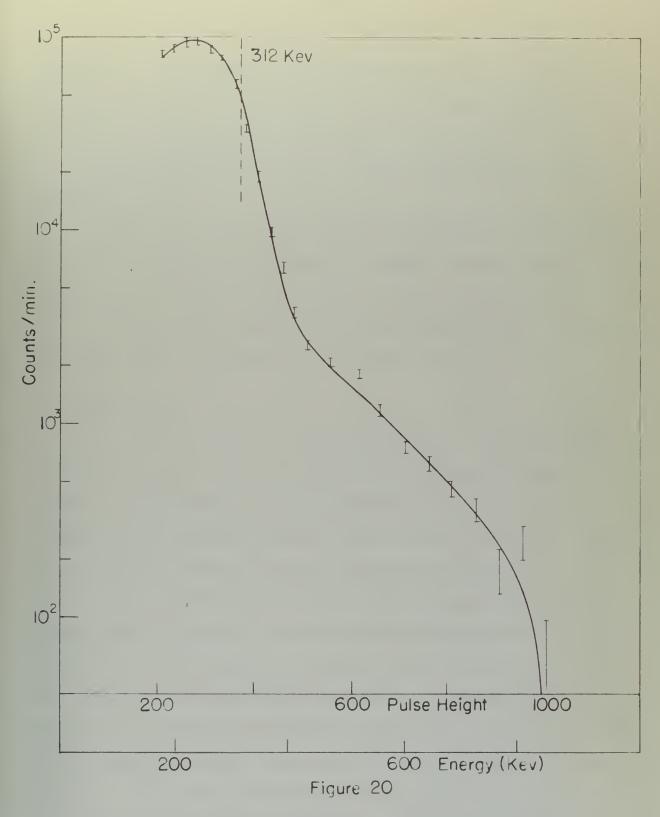
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DIFFERENTIAL PULSE HEIGHT DISTRIBUTION for In 115 from THICK FOIL SHOWING 312 Kev CONV. ELECTRON LINE and 830 Kev BETA DECAY (Energy calib. from Fig. 6)



excitation. A spin change of 2 with parity change could be provided by an ℓ = 1 or an ℓ = 3 neutron; if this is the case, neutrons will excite this level strongly. On the other hand, a spin change of 3 with parity change requires an ℓ = 3 neutron, giving a small probability.

Unfortunately, it was not possible to determine the cross section for neutron excit tion in this ration. To refere, no choice can be made between those two, and both spins are given on Figure 19.

For the case of the higher levels, many spin and varity choices will give results consistent with the observed excitation curves.

FXPERIMINAL SETU

The indian samples were in the form of fells 8 mils thick and 3 cm. diameter. These feils were irradiated for h hours with monomorphism from the Li⁷(p,n) reaction using a 20-keV thick target on the Reckefeller generator. Ifter irradiation, the feils were counted in the scintillation counter, described in Chapter III.

Such a long exposure, necessitated by the long lifetime of the isomer, introduces several problems into the determination of the excitation curve. It is impossible to keep the neutron flux constant over the exposure without sacrificing a portion of it to provide a range of adjustment. If the flux changes during this period, a simple integration of the neutron flux is not sufficient for the det reinstion of the cross section. At the time these measurements were made, a continuously recording neutron-flux indicator was not available. It was therefore nec-

Industriality it was not provided as demands the cours desirable.

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Description of the property contractions by the last last three distriction of the centdecise, relaxations covered contract the few terminations of the central decision of the central decision of the property or the property of the property of the property of the property of the contract of the property of the prop essary to obtain several points on the excitation curve during the same irradiation, thus determining a section of the curve. These sections could then be joined by overlapping.

Another factor to be considered is the availability of generator time. If each point on the excitation curve is determined separately, the exposure time required becomes prohibitive.

For these reasons, advantage was taken of the dependence of the neutron energy on angle, and seven foils were exposed simultaneously at angles ranging from 0 to 75 degrees. Figure 21 shows the foil arrangement for exposure.

In order to obtain sufficient activity in the foils, it was necessary to place the foils so that the marest edge was only I em. from the neutron source. At this distance, the mergy resolution was preserved by rolling the foils into small cylinders about 3 mm. in dismeter and mounting them radially about the source.

The effect of capture of thermal neutrons was minimized by counting the side of the foil rolled inside. All of this capture activity is located in the very thin layer which was octside; so that most of the foil is shielded from thermal neutrons.

After irradiation the foils were counted in rotation six to eight times over a period of about 8 hours, starting with the lowest activity to separate the 5k-minute capture activity from the h.5-hour activity of In 115*. To increase the ratio of In 115* to In 116 counts, advantage was taken of the energy sensitivity of the scintillation counter. The

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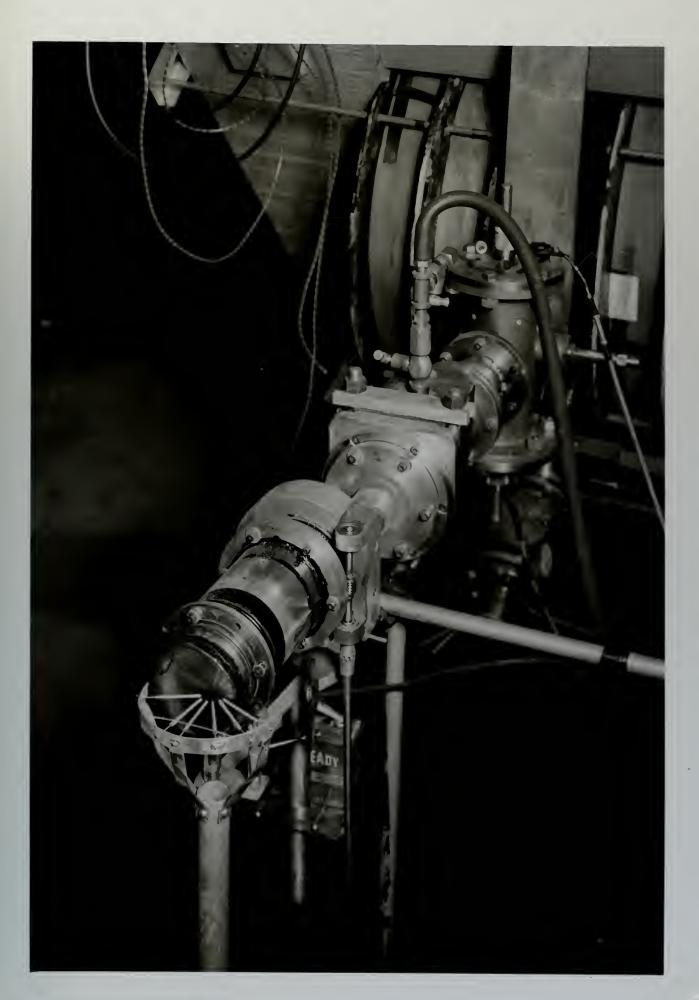
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Figure 21. Foil arrangement for the indium activation showing the foils rolled on the wire of the spider.

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levels of the differential discriminator were set to record counts corresponding to an energy between 150 and 150 keV. This includes most of the counts from ${\rm In}^{115*}$ (see Figure 20) but includes only part of the counts from ${\rm In}^{116}$ (E_{max} = 850 keV for the beta-particle emitted).

An attempt to separate the two activities was made by comparing the number of counts above and below the pulse height corresponding to 450 keV, thus using the difference in shape of their puls -height distributions. However, it was found that the statistical uncertainty in this method made it impracticable.

TREATMONT OF THE DATA

In order to take proper account of all points making up the decay curve of a given foil, it was considered desirable to compare the experimental curve with a theoretical decay curve. Since the construction of such a theoretical curve presupposes a knowledge of the relative amounts of the two activities present, a curve to include all cases had to be found.

All the experimental curves were plotted with the same scale on the same type of semilogarithm graph paper. A master decay curve was then constructed, also using the same scale, on a large sheet of paper starting with an assumption of the shorter-lived activity being 100 times that of the longer-lived and continued until the shorter-lived activity had decayed to 1/100 of the longer-lived. It is evident that, if the experimental curve is made up of a combination of the two activities within these limits, it must fit the master curve over some

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interval. By superposing the experimental curve on the master and by sliding the former to find the best fit, each point could be given its proper weight. To permit determination of the amount of such ctivity, two lines were drawn on the master corresponding to the contribution of each activity to the total curve. It was then a simple matter to read off the amount of each activity at any time and in particularly at the time corresponding to t = 0 on the experimental plot.

In this investigation, the 105-minute isomeric activity in Inll3 was neglected. That this would be possible was predicted from the relative abundance (1.2 percent for Inll3, compared with 95.8 percent for Inll5). The decay curves obtained justify this assumption, inasmuch as the combination of the 5h-minute and the 4.5-hour activities were sufficient in all cases to explain their shape.

The accuracy of the method described above was high enough so that the uncertainty in the determination of the In 115* activity was estimated to be less than 10 percent.

Since the neutron flux is anisotropic, the various activities must be corrected as a function of the exposure angle. Taschek and Remarkinger have shown that the flux is not even isotropic in the center-of-mass coordinates for the Li⁷(p,n) reaction because of the resonance in the compound nucleus. The flux as a function of angle must therefore be measured. Taschek and Hermandinger's measurements do not extend into the region of interest in this work. In conjunction with Willard, the neutron flux as a function of angle from 0 to 90

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degrees was measured for proton energies from the Li⁷(p,n) threshold to 3.75 lev using the long counter described in Chapter III. The scattered neutron background was determined by use of a paraffin cone to shadow the counter. The results of these measurements are reported by willard⁵⁵, and are plotted here in Figure 21a.

Each irradiation of folls covered an energy interval of from 300 to 500 kev. The intervals were chosen so that they overlapped approximately half of their extent. Each segment of the In^{115*} excitation curve determined in the interval was corrected for the anisotropy of the flux and energy resolution and was then normalised to the preceding segment. This technique involves a propagation of errors but does not affect the location of energy levels found by means of the curve.

Figure 22 shows the complete curve.

DISCUSSION OF THE RESULTS

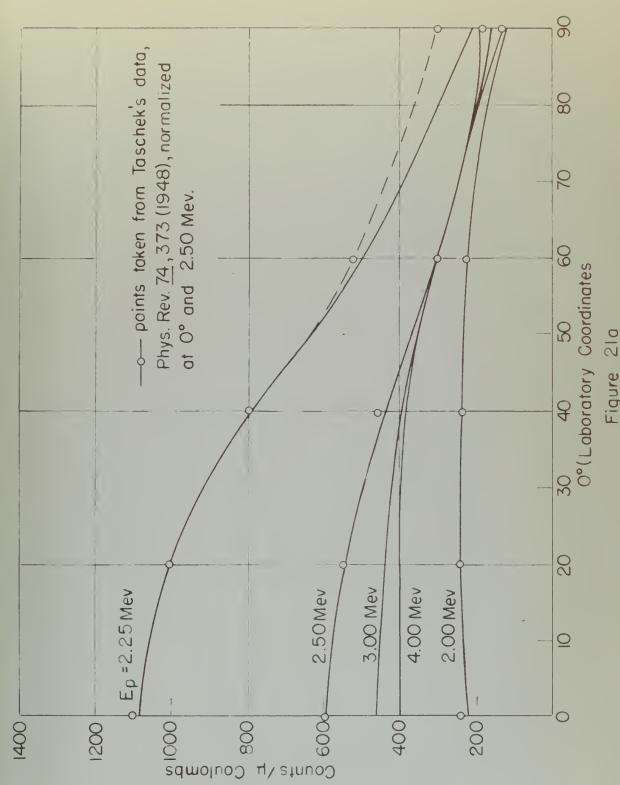
In spite of the scattering of the points on the excitation curve (Figure 22), the presence of three levels is shown at 600 keV, 960 keV, and 1.37 keV with ressibly a fourth level at 1.75 keV. The uncertainty in these energy assignments is estimated to be of the order of h0 keV. This uncertainty, however, is considered hardly sufficient to bring the 960 keV and the 1.37 keV level into agreement with Miller and Waldman's values of 1.02 and 1.h2 keV for quantum-excited levels. The correspondence is close enough to leave little doubt that the same levels are being excited and suggests that the energy calibration for one of the experiments is in error. Unfortunately, there is not a third accurate

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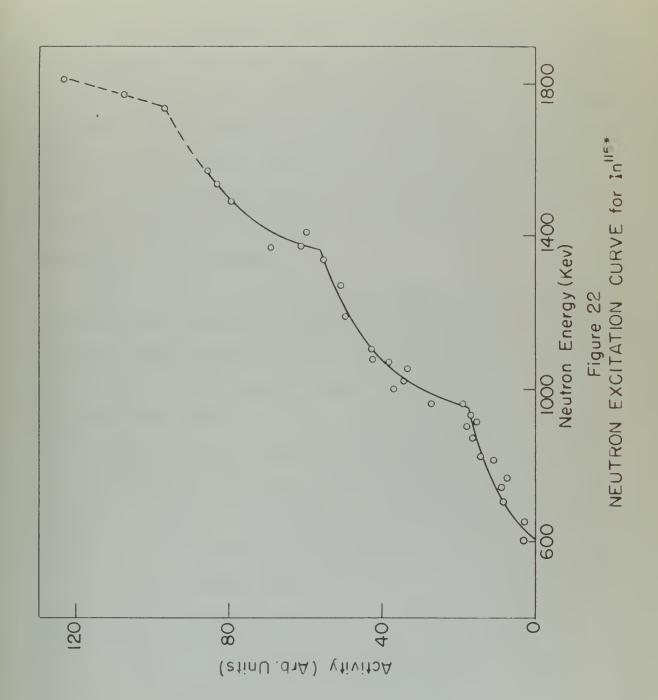
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RELATIVE DIFFERENTIAL CROSS SECTION for the Li7(p,n)Be7 REACTION







determination of eith r of these levels for comparison. From the reproducibility of the calibration of the lockef liber gener tor, it is difficult to see how it could be in error by the amount surgested above.

Probably the greatest source of error in the individual points on the excitation curve is due to the stringent requirements on positioning the foils. Since the point at which the foil may be considered concentrated for the irradiation is only 2.1 cm. from the target (found by integrating an inverse square activity over a circular foil), a 2-am. error in the mounting of the foils or in determining the point at which the proton beam strikes the target results in 20 percent error in the induced activity.

the location of the beam cannot be observed after the target has been installed so that it is impossible to know wheth r the beam has wandered slightly during the irradiation. This error can be corrected for, however, because foils were exposed at angles on both sides of 0 degrees. It was then assumed that the beam actually hit the target at such a point that the activities were symmetrical about 0 degrees.

The placement of the individual feels on the circumference of a circle with its center at the target could not be checked in such a simple fashion and is no doubt the reason for the spread in the points.

In further investigation of this problem, it is suggested that a thicker target (of the order of 50 kev) be used and the fails be mounted further from the target, even at the expense of reducing the activity below that obtained in this investigation.

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VI. CONCLUSIONS AND SUCCESTIONS FOR PURPLE INVESTIGATION

SUBMEN OF PESULES

Noutron excitation reveals the existence of energy levels in ${\rm Au}^{197}$ at 1.14 Nev and 1.14 Nev. It is found that the metastable state at 540 keV can be excited directly by ℓ = 3 neutrons. The spin of this level is shown to be 11/2 with parity opposite that of the ground state. This fixes the spin of the 270-keV level to which the metastable level decays at 5/2 with parity also opposite that of the ground state.

Since the 1.11-Mov level is also excited by photons, its spin is shown to be 7/2 or 9/2 with parity opposite to that of the ground state.

An analysis of the data for direct production of the metastable state shows the cross section to be reasonably in agreement with that predicted by the continuum theory for the production of the compound nucleus by an $\ell=3$ neutron.

The levels found in In 115 by neutron emitation are at 600 keV, 960 keV, and 1370 keV. The latter two are also found by quantum excitation, but the quantum values are slightly higher (8 percent). An analysis shows that the metastable level has a spin equal to 1/2, and the 600-keV level, a spin of 5/2 or 3/2. Both of these levels have a parity different from the ground state.

FUNTIER INVESTIGATION OF Au 197=

Because of the disagreement between energy levels obtained in the investigation and those obtained by Wiedenbeck using thick-target x-rays,

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it would seem of great value to redetermine the quantum excitation curve using x-rays from a thin target. Determination of the relative probability for exciting the various levels in this fashion would indicate possible spin and parity values for some of the higher excited levels.

An extension of the neutron excitation curve to higher energies would probably reveal the presence of other levels. A logical choice for the neutron source in such a study is the d-d reaction which will yield from 2.5-Nev to 7-Nev neutrons with the Rockefeller generator. The use of deuterons as the particle accelerated has the disadvantage of giving very high backgrounds of moutrons even though the beam is not striking the target. This is partly because so many of the douteron reactions have low thresholds. In addition, deuterons in the beam are imbedded in the metal a distance corresponding to their range. Then the accelerating voltage is raised slightly, this forms an effective target for the d-d reaction.

However, if a counter for the gold activity can be found that is insensitive to this background or if it is possible to cut off the beam farther from the counter, it will be possible to extend the results of this investigation.

FURTILE INVESTIGATION OF IN-115*

Better results could be obtained for the emitation of In 115° if the positioning error could be reduced by exposing the foils at a greater distance from the target. In Chapter V, it was suggested that resident contract and indirectables of refer decorpts come blace at several expense pulsar recent which follows not published at a final follows and published by the follows and published by the follows and the follows and the follows are the follows.

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this be accomplished by use of a thicker lithium target. In addition, the beam current available from the Rockefeller generator has been increased by a factor of 3 or 4 since the exposure were made. It is felt that, with a 50-kev lithium target and the present beam current on the order of 5 microsspores, sufficient activity could be obtained in the foils when they are mounted with the nearest edge on the order of 6 cm. from the target.

motor that operates a continuously recording motor, such as an Esterline-Angus pen recorder, the neutron flux on the foils would be determined as a function of time. This, plus a measurement of the efficiency of the In
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Exercise a continuously recording motor, such as an Esterline-Angus pen recorder, the neutron flux on the foils would be determined as a function of the efficiency of the In
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Exercise a continuously recording motor, such as an Esterline-Angus pen recorder, such as an Esterline-Angus pen recorder, the neutron flux on the foils would be determined as a function of the eross section of the efficiency of the eross section with the theoretical value will remove the ambiguity in the spin of the 600-key level.

INVESTIGATION OF OTHER ISOTOPES

Several other isotopes (see Table I) lend themselves to an investigation similar to that made on gold and indium. All may either be treated according to the technique used for gold or that used for indium.

Rhodium has the advantage of being composed of 99.9 percent of the isotope containing the metastable state, Rh¹⁰³. The excitation by x-rays has been accomplished by Tiedenbeck, as reported in Chapter I. The metastable state is only of the order of 60 kev above the ground state, but radiation of this energy can be detected easily by a scintil-

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lation counter. Its half-life of 15 minutes requires that any extensive investigation be carried on in a fashion similar to that used for indium.

The excitation of Cdlll by Medenback has also been reported in Chapter I. This state has an energy of 119 kev with a half-life of 10.7 minutes. In addition, a metastable state in Cdll3 has been found by N elmholz and McGimnis with a half-life of 2.3 minutes. The cross sections for both of these activities are expected to be small because they occur in isotopes with only about 12 percent relative abundance. However, there is no activity expected that would confuse the results, and their lifetimes are such that counting may be made at some distance from the generator, resulting in low background.

Another possible choice for investigation is Ba¹³⁷ with a 158-second half-life. Unfortunately, this isotope is also low in relative abundance, being Il percent. There have been no investigations of the excitation curve reported for this state.

Nafnium has a 19-second, 200-kev activity assigned to either 18177 or Hf179. The former is 13 percent abundant and the latter, 1k percent. Although it may prove possible to lower the background during the counting period by removing the accelerator voltage, it is more likely that the beam will have to be cut off by a flap valve as in the case of gold. If this is true, the discussion for wolfram given below also applies to hafnium.

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Last among the logical choices is W¹⁸³. This isotope of wolfram could be investigated with the technique used for gold, since its half-life is 5.5 seconds. The isotopic abundance of 1½ percent will result in a low cross section in all probability. Unlike barium and cadmium, however, the lifetime is so short that the voltage cannot be removed from the accelerator, and backgrounds on the order of those obtained for gold will be encountered. If the same cross section per nucleus is assumed for 183 as was found for Au¹⁹⁷, the activity expected is so low that the determination of the wolfram curve is a marginal case.

In summary, if several rhedium foils could be obtained, Rh¹⁰³ would seem the best choice for future investigation, followed by the two cadmium isotopes and Ba¹³⁷. Hafnium and wolfram appear to pose a very difficult problem with the present neutron source strength.

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VII. ACKNOVLINGHENTS

I wish to thank Dr. Clark Goodman for his advice, interest, and constructive criticism during the progress of this research and the preparation of this thesis. Drs. Victor F. Weisskopf and Marvin L. Goldberger contributed many valuable suggestions on the theoretical aspects of the problem. I am also indebted to Dr. William W. Euschwer for helpful discussions about this research.

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by appreciation is due Mrs. Mary E. White and Mrs. Charles Rows, Jr. for their excellent work in the preparation of the manuscript.

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Education:

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Iowa State Teachers College, Cedar Falls, Iowa, B. A., 1942

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Associations:

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